Uptake of SO_2 as a Function of pH

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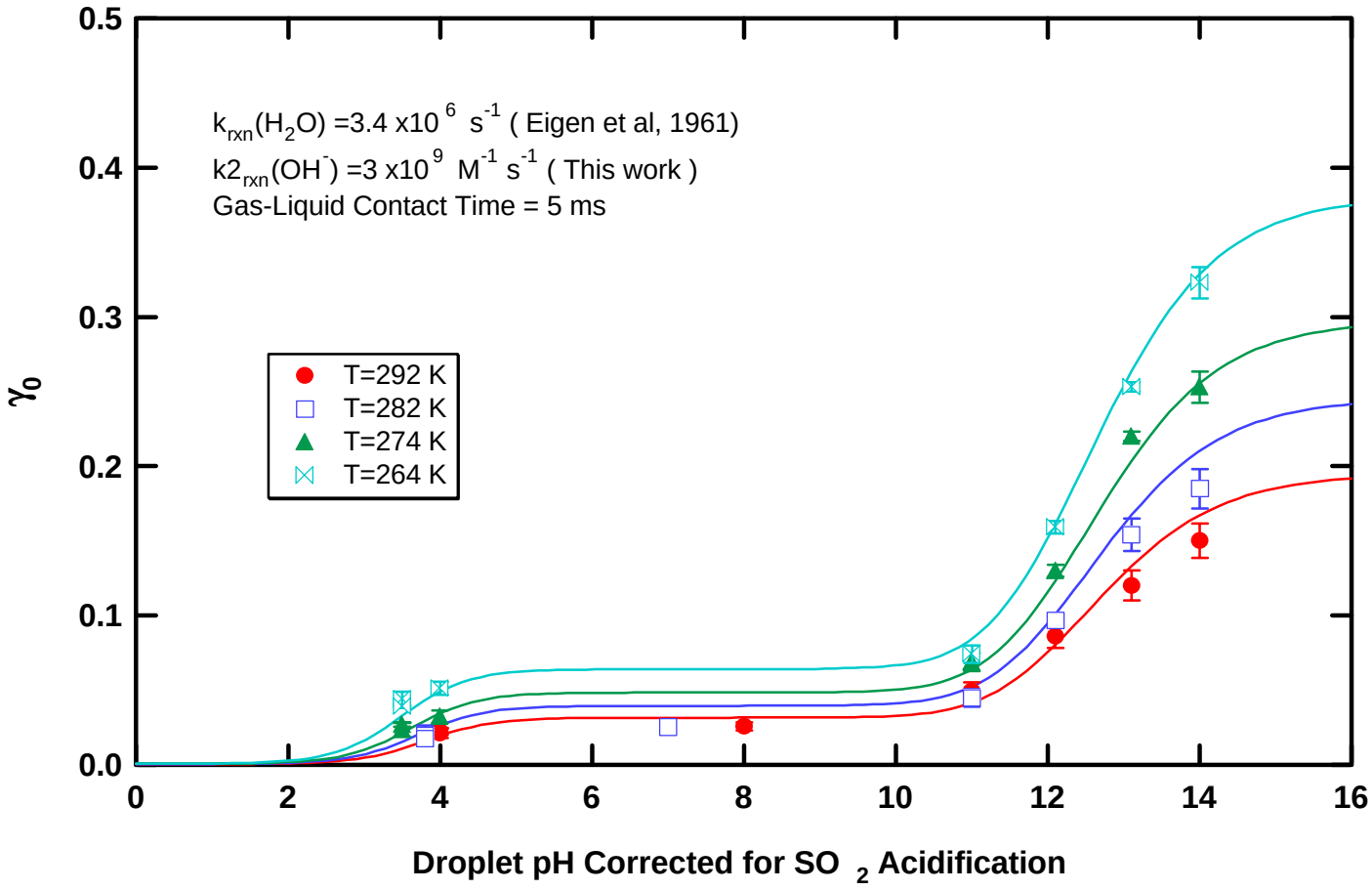
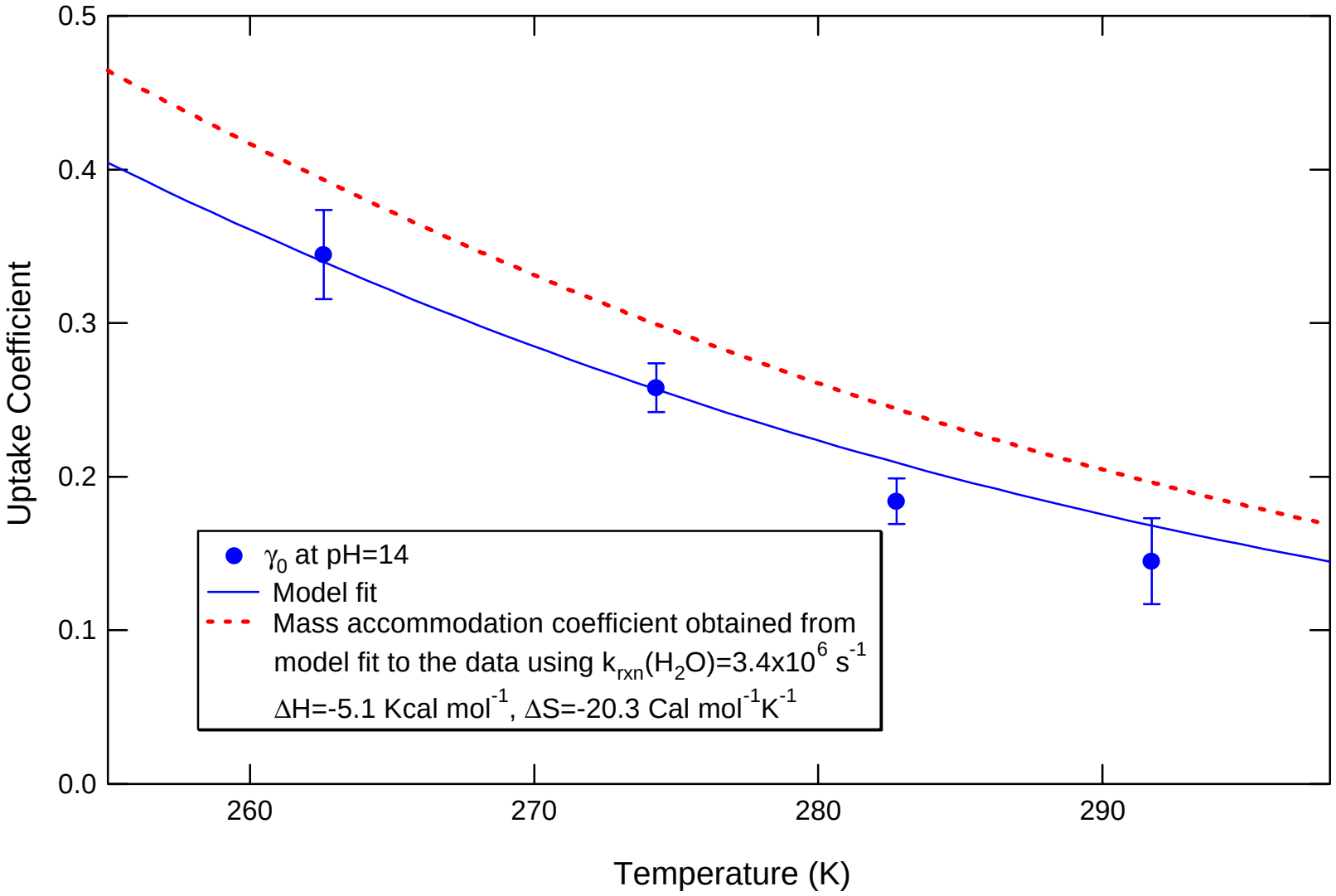
Using the droplet apparatus, earlier SO_2(g) uptake studies were extended to higher pH up to pH = 14. Measurements were conducted as a function of gas-liquid contact time (1.8 ms to 7 ms), droplet temperature (T = -10 ^{o}C, 0 ^{o}C, 10 ^{o}C, and 20 ^{o}C). To confirm gas phase diffusion treatment two droplet-generating orifices of diameters 64 μm and 28 μm were used.

SO_2 Uptake as a Function of pH and Temperature



Uptake coefficient γ_o is corrected for gas-phase diffusion

SO_2 Mass Accommodation Coefficient as a Function of Temperature



Results:

- Results at lower pH are consistent with earlier studies confirming the existence of a surface complex.

- The at higher pH the uptake continues to rise most likely due to a direct reaction $SO_2 + OH^-$ -----> HSO_3

- The second order reaction rate is $\mathbf{k_2 = 3x10^9\ M^{-1}\ s^{-1}}$.

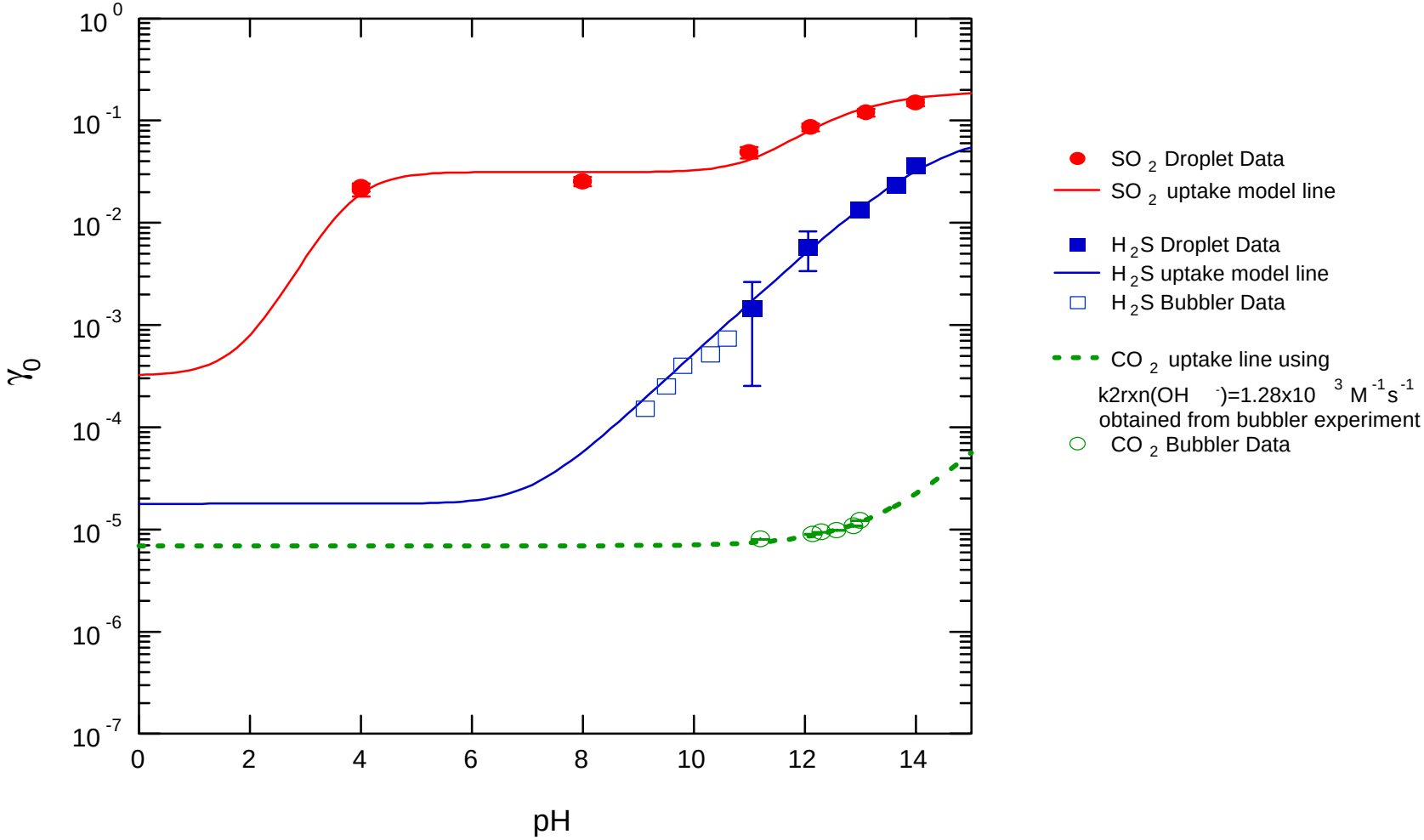
- As pH increases uptake rises toward the value of the mass accommodation coefficient α.

- The mass accommodation coefficient is consistent with the clustering model and can be expressed as

$\alpha/(1 - \alpha) = \exp(-\Delta G/RT)$

With $\Delta H = -5.1$ kcal mol^{-1} and $\Delta S = 20.3$ cal mol^{-1} K^{-1}
At -10^oC $\alpha = 0.36$

Uptake of SO_2, H_2S and CO_2 as a Function of pH at 291 K and Gas-Liquid Contact time 5 ms



Interactions of SO_2, H_2S, CO_2 with OH^-

Summary of Data:

at 291K

SO_2: $k_2(OH^-) = 3.0 \times 10^9\ M^{-1}s^{-1}$

H_2S: $k_2(OH^-) = 1.8 \times 10^9\ M^{-1}s^{-1}$

CO_2: $k_2(OH^-) = 1.3 \times 10^3\ M^{-1}s^{-1}$